

Actinide (n,f) cross sections using the surrogate technique



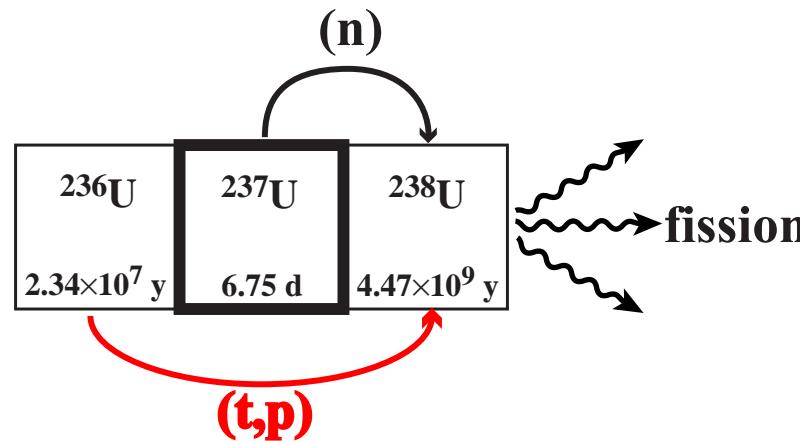
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Motivation, problem, and solution

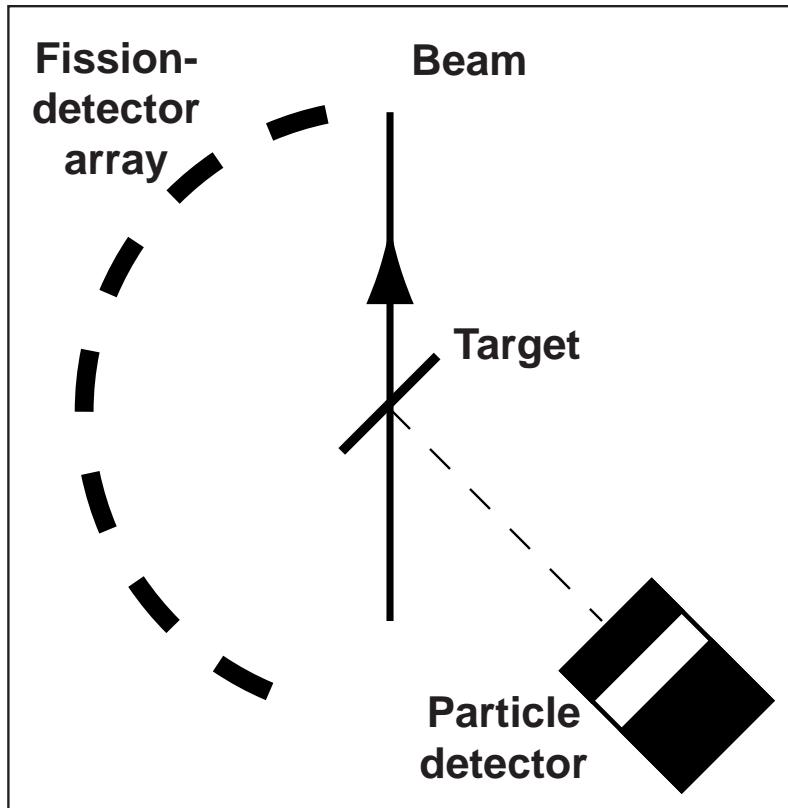


- Motivation: RadChem network requires complete set of cross sections.
- Problem: (n,f) on many actinides cannot be measured because target lifetime is too short (e.g. $T_{1/2}(^{237}\text{U}) = 6.8 \text{ d}$).
- Solution:
 - Use surrogate reaction, e.g. (t,p) to populate the same compound nucleus (^{238}U) as (n,f)
 - Use model to compensate for differences between (t,p) and (n) reactions (angular momentum, parity)



Surrogate technique
– enables otherwise unobtainable cross sections

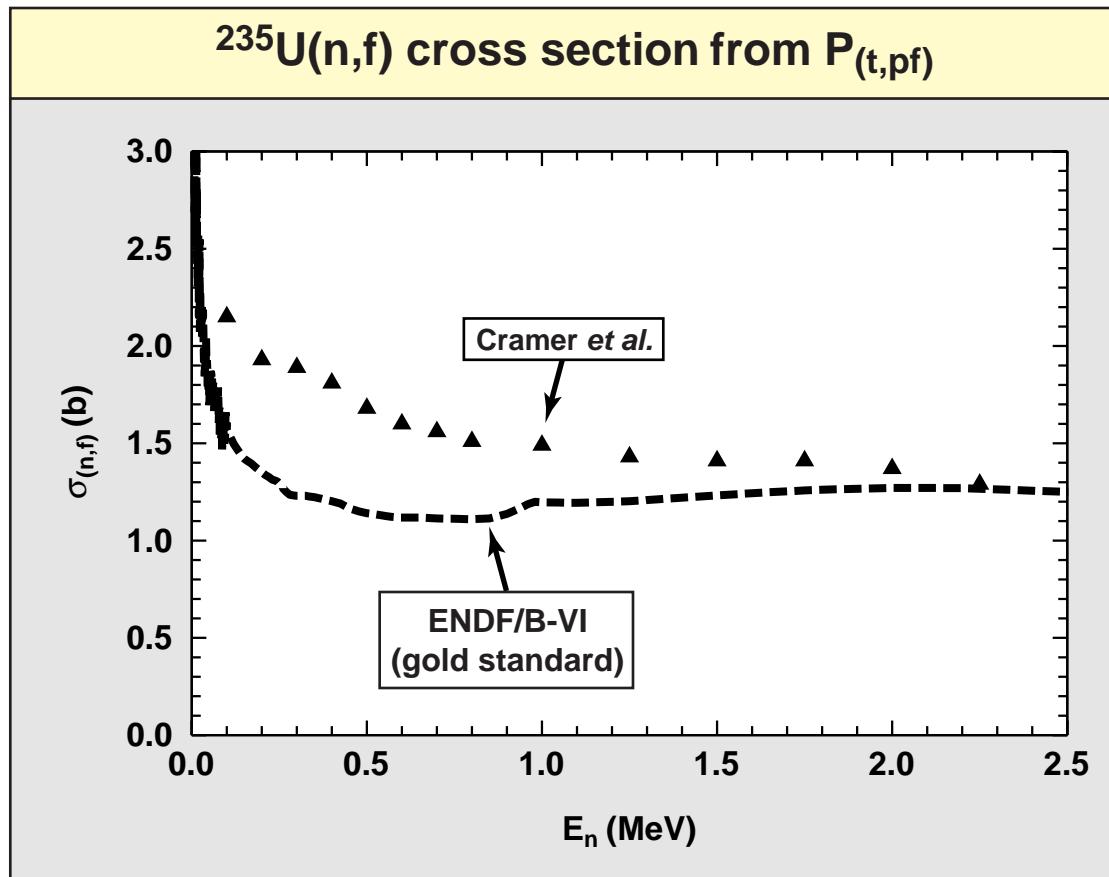
Experimental setup for (t,pf) at LANL Van de Graaff



- Measure (t,p):
 - E = proton energy
 - N_p = # protons
 - N_{pf} = # proton-fragment coinc.
- Extract:
$$P_{(t,pf)}(E) = \frac{1}{2} \times \frac{4\pi}{\Delta\Omega} \times \frac{N_{pf}(E)}{N_p(E)}$$
- Backgrounds:
 - ¹²C and ¹⁶O in target
- Who, Where, and when:
 - Back, Britt, Cramer, Wilhelmy, ...
 - Los Alamos Van de Graaff
(late 60's to mid-70's)

Large database of measured fission probabilities:
– 62 nuclei from Th to Es

$^{235}\text{U}(n,f)$ - Validation: (t,pf) to (n,f) in a simple approach



- Cramer et al. (1970)

$$\sigma_{(n,f)}(E_n) = \sigma_{CN}(E_n) \times P_{(t,\text{pf})}(E_x)$$

Previous calculations were hampered by:
- Lack of treatment of J^π
- Inadequate modeling of σ_{CN} for low E_n

From (t, pf) to (n, f): more sophisticated approach



Key Concepts:

- Measured fission prob.
 - contains contributions from all J^π
- Use model to extract J^π contributions
- Assume formation and decay are separable

Basic technique:

Measure Calculate Fit model

$$P_{(t, pf)}(E_x) = \sum_{J^\pi} P_{(t, p)}(J^\pi) \times P_f(E_x, J^\pi)$$

Reuse

$$\sigma_{(n, f)}(E_n) = \sum_{J^\pi} \sigma_{CN}(E_n, J^\pi) \times P_f(E_x, J^\pi)$$

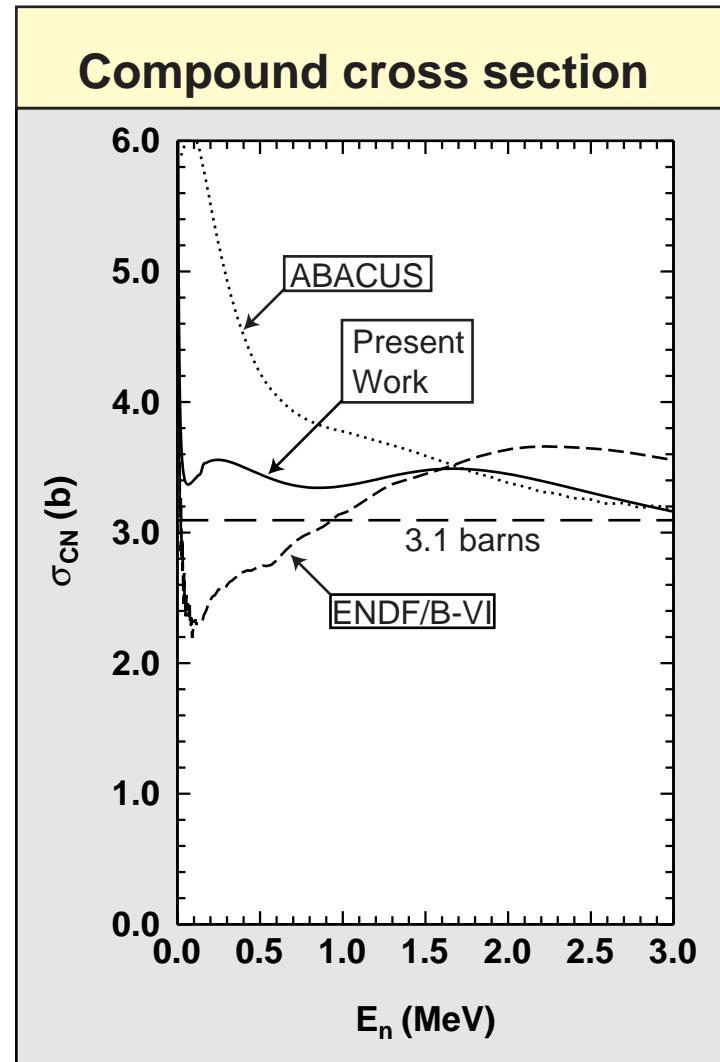
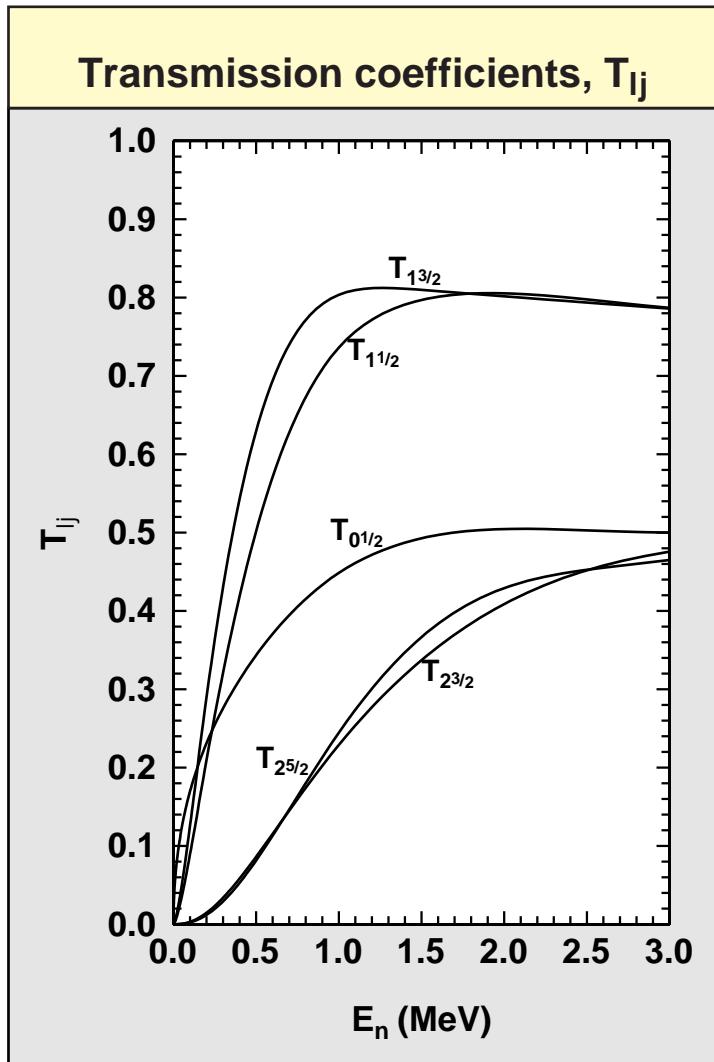
Deduce

Calculate

Key ingredients:

- Neutron population $\Rightarrow \sigma_{CN}$
- Direct-reaction population $\Rightarrow P_{(t, p)}$
- Fission model $\Rightarrow P_f$

Neutron-induced population

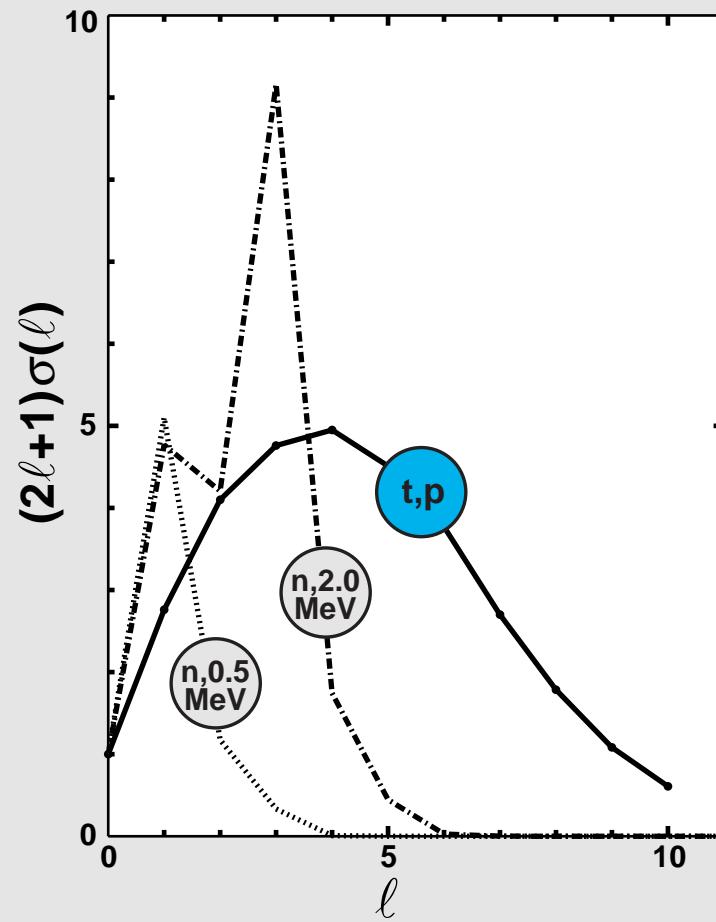


New transmission coefficients (F. Dietrich)
- more realistic

Direct-reaction population

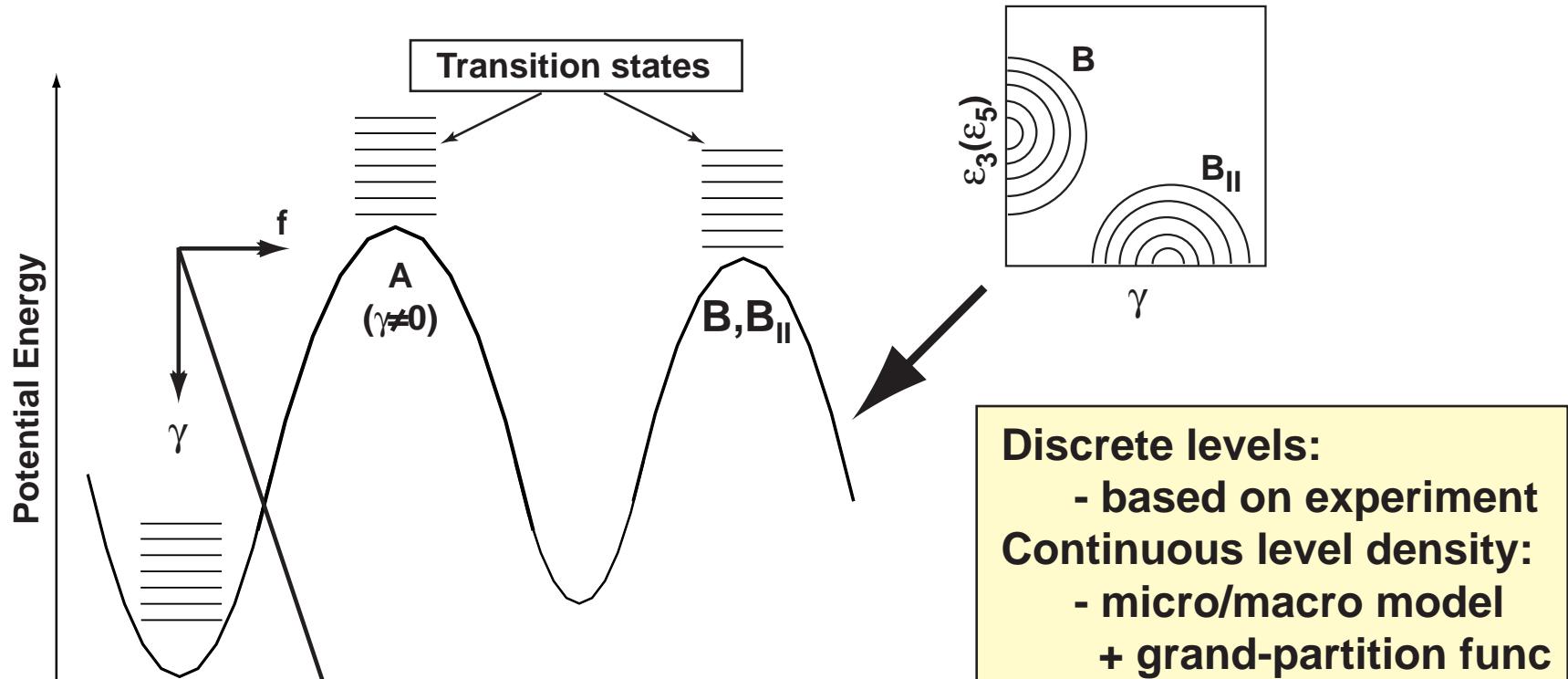


Angular momentum dependence



Populations via n-capture and (t,p) differ
— Must be modeled

Bohr Fission Model



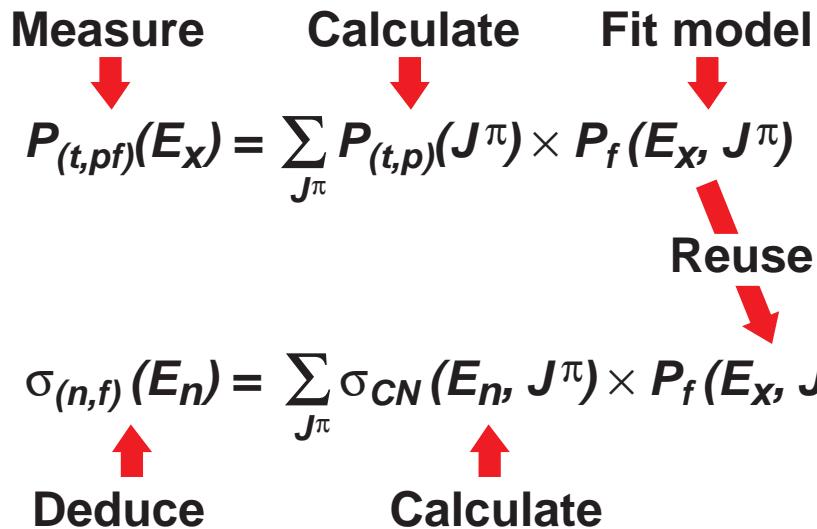
Procedure:

– adjust barrier A & B heights to fit measured $P(t, pf)$

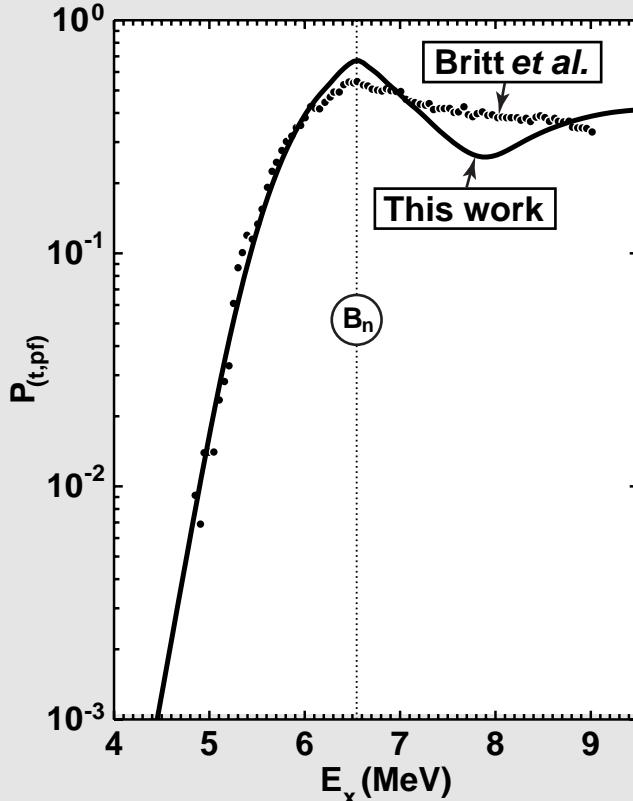
Proof of principle I: $^{235}\text{U}(n,f)$ from $^{234}\text{U}(t,pf)$



Basic technique:

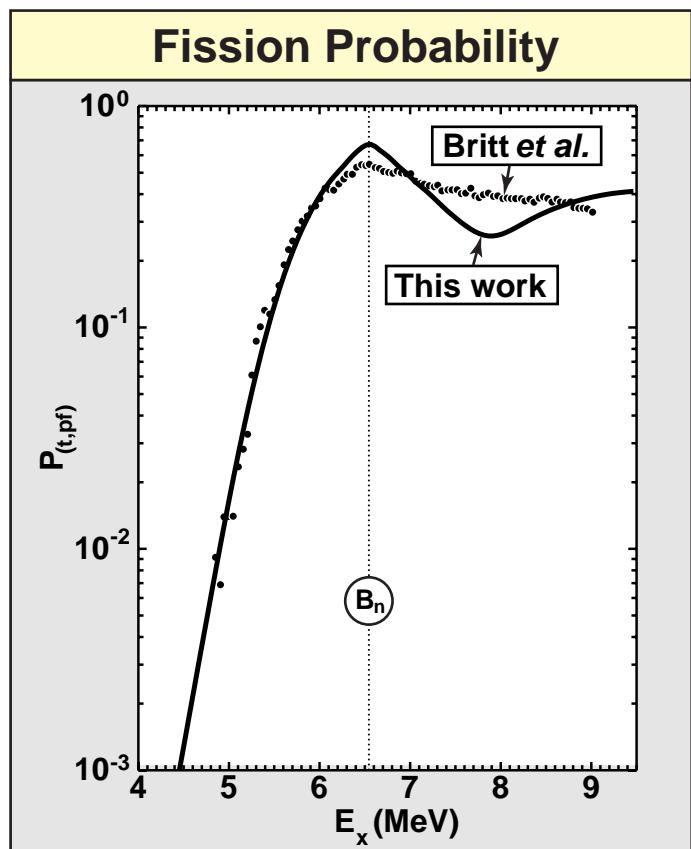


Best Fit to Fission Probabilities

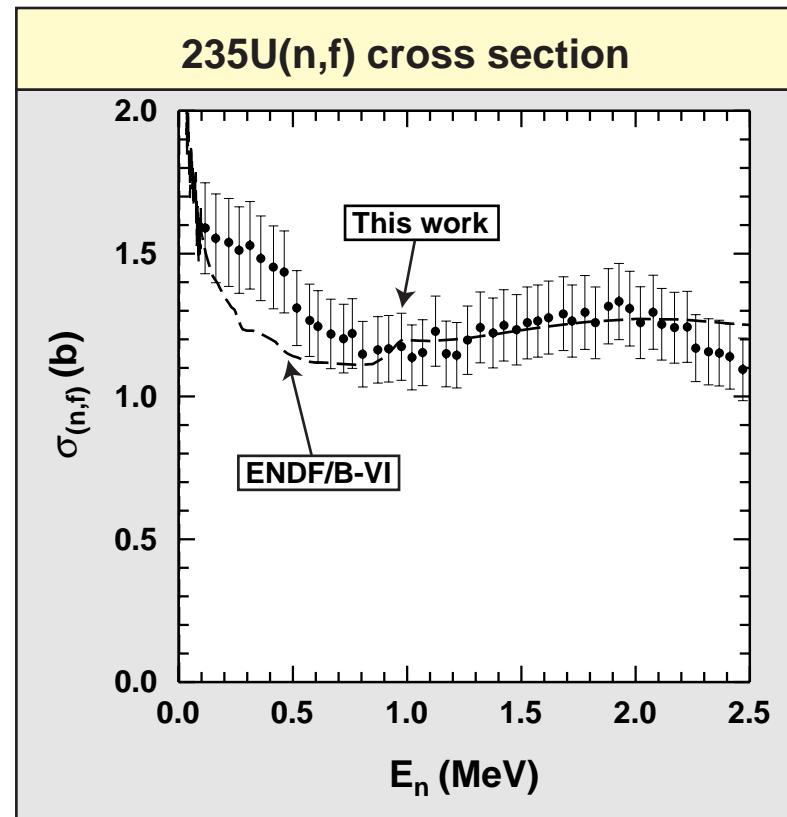
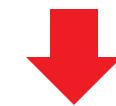


- Fit is not perfect
 - Model is over-constrained
 - Relative $P_f(E_x, J^\pi)$ are robust
- Renormalize to $P_{(t,pf)}$ data

Proof of principle II: $^{235}\text{U}(n,f)$ from $^{234}\text{U}(t,pf)$

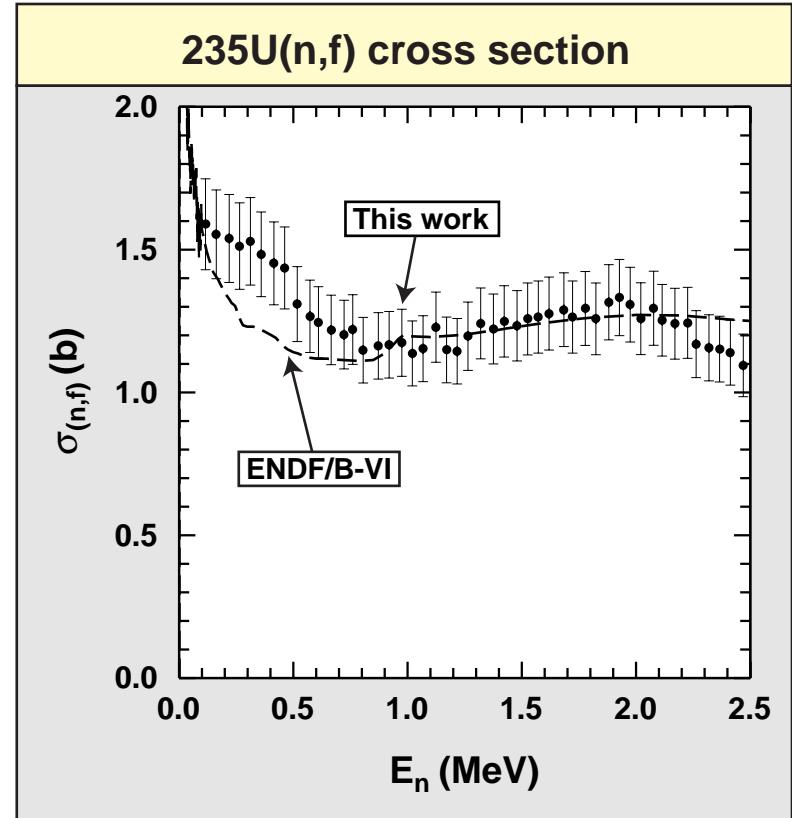
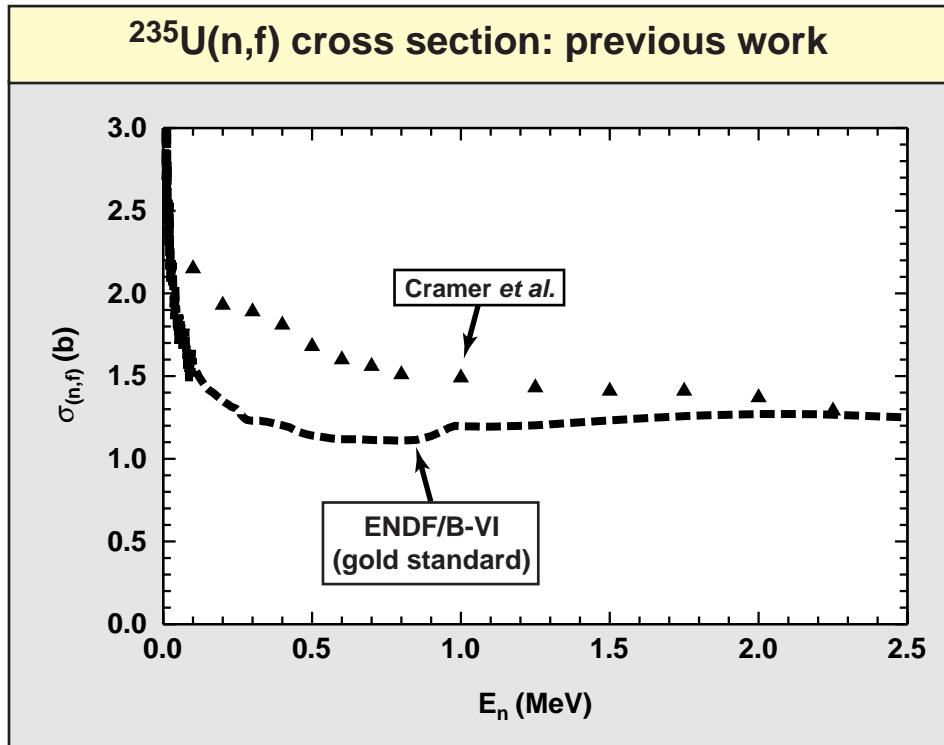


$$\sigma_{(n,f)}(E_n) \times \frac{P_{(t,pf)}^{(\text{expt})}(E_x)}{P_{(t,pf)}^{(\text{calc})}(E_x)}$$



$\sigma_{(n,f)}$ is good to
 - 20% below ≈ 1 MeV
 - 10% above ≈ 1 MeV

Comparison with previous estimate



New surrogate results improve significantly on old results

$(t, pf) \Rightarrow (n, f)$: Systematic deviations



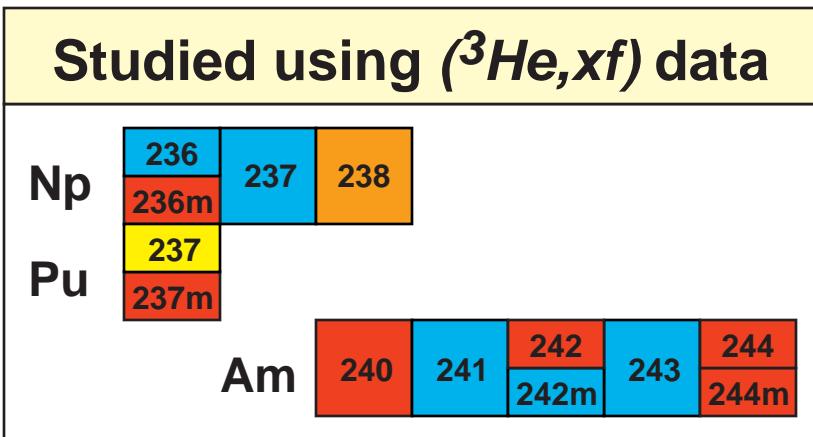
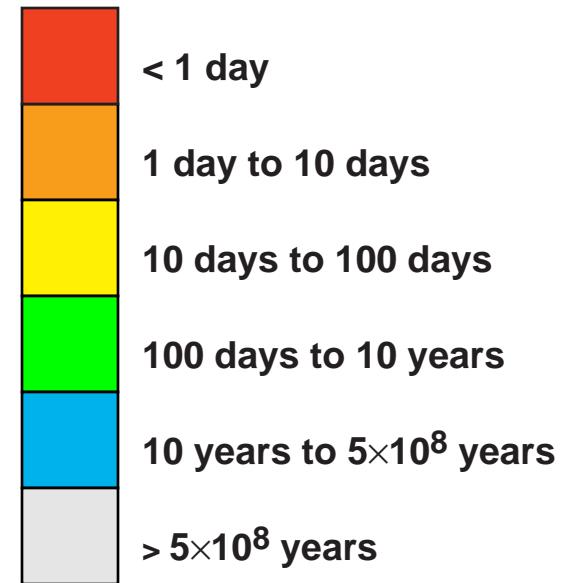
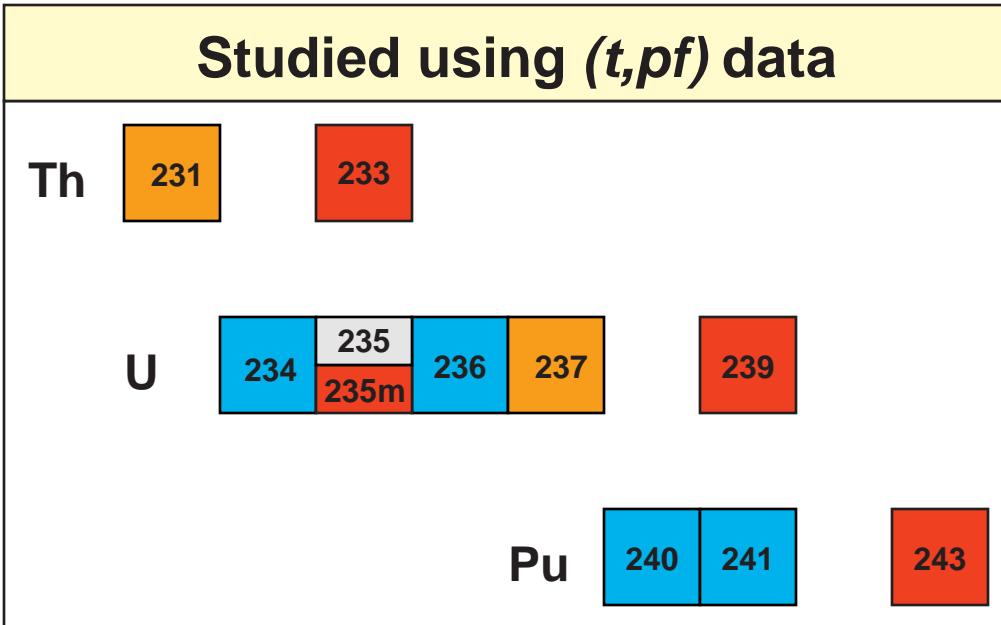
- Compare to known cross sections for $E_n = 1\text{--}3 \text{ MeV}$

Neutron Target	Surr. $\langle\sigma(n,f)\rangle$ (barns)	ENDF $\langle\sigma(n,f)\rangle$ (barns)	Relative dev. (%)
^{234}U	1.37	1.45	-5.5
^{235}U	1.23	1.23	0.0
^{236}U	0.87	0.77	+13.5
^{240}Pu	1.51	1.61	-6.2
^{241}Pu	1.40	1.64	-14.6

Surrogate cross sections averaged over 1-3 MeV are consistent with (n,f) measurements to $\pm 10\%$



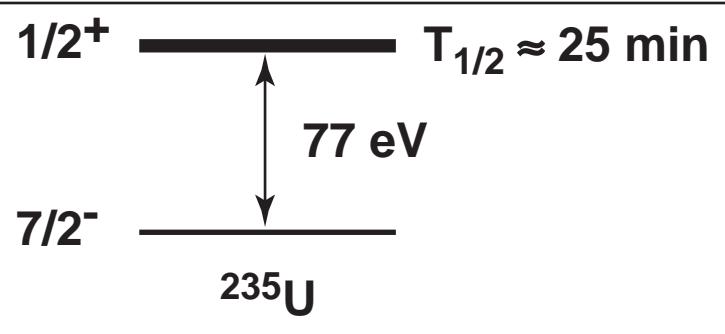
(n,f) cross-section results to date



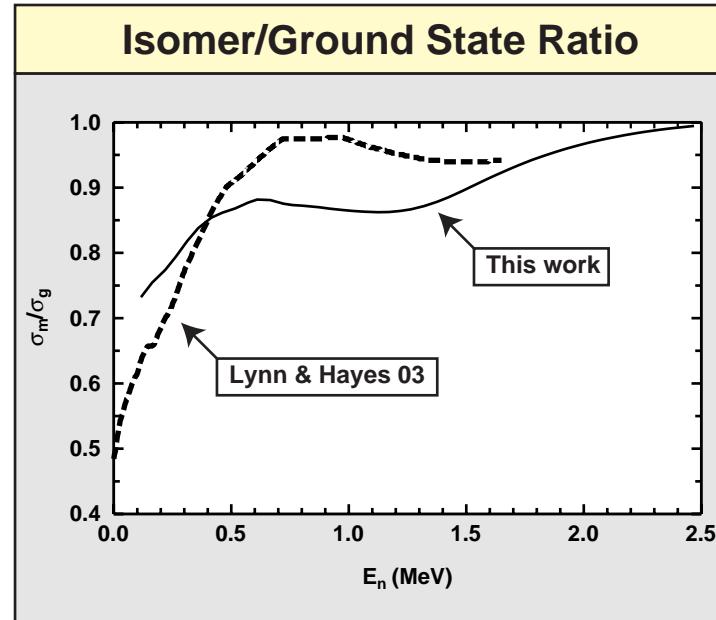
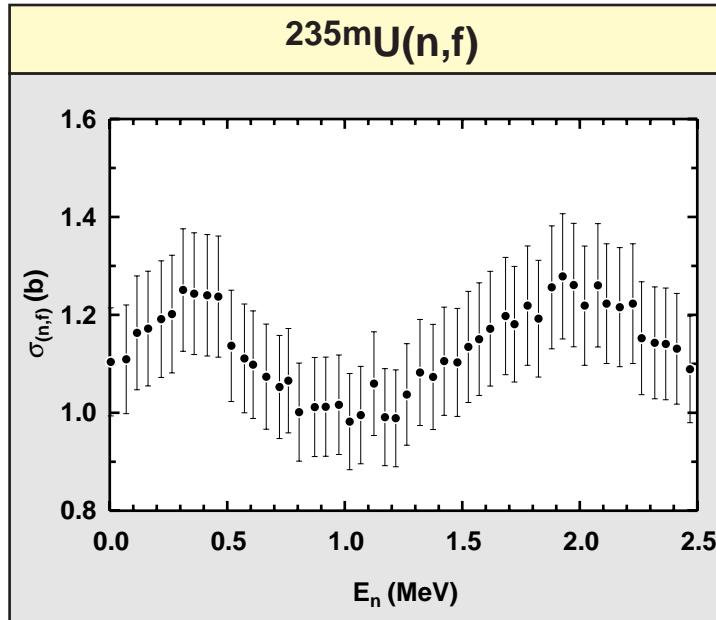
Results written up in

- Younes & Britt, PRC 67, 024610 (2003)
- Younes & Britt, PRC 68, 034610 (2003)
- Many LLNL internal reports...

Application: The $^{235m}\text{U}(n,f)$ cross section



- Requires only incremental effort:
 - Change target J^π in n-capture calc.
 - $P_f(E_x, J^\pi)$ same as in ground-state calc.
 - σ_g well reproduced \Rightarrow reliable σ_m

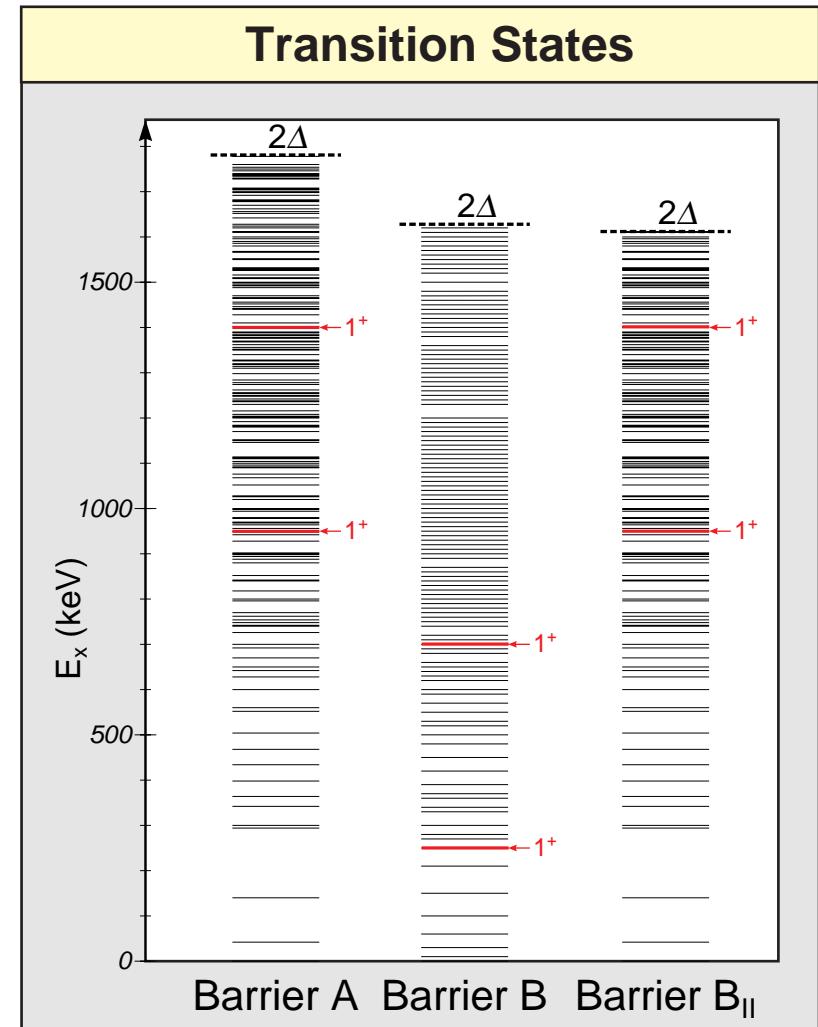


- Similar results found by Lynn and Hayes: PRC 67, 014607 (2003)
- $\sigma_m/\sigma_g < 1$ for low E_n : why?

Low-spin transition states have big effect on (*n,f*) cross section

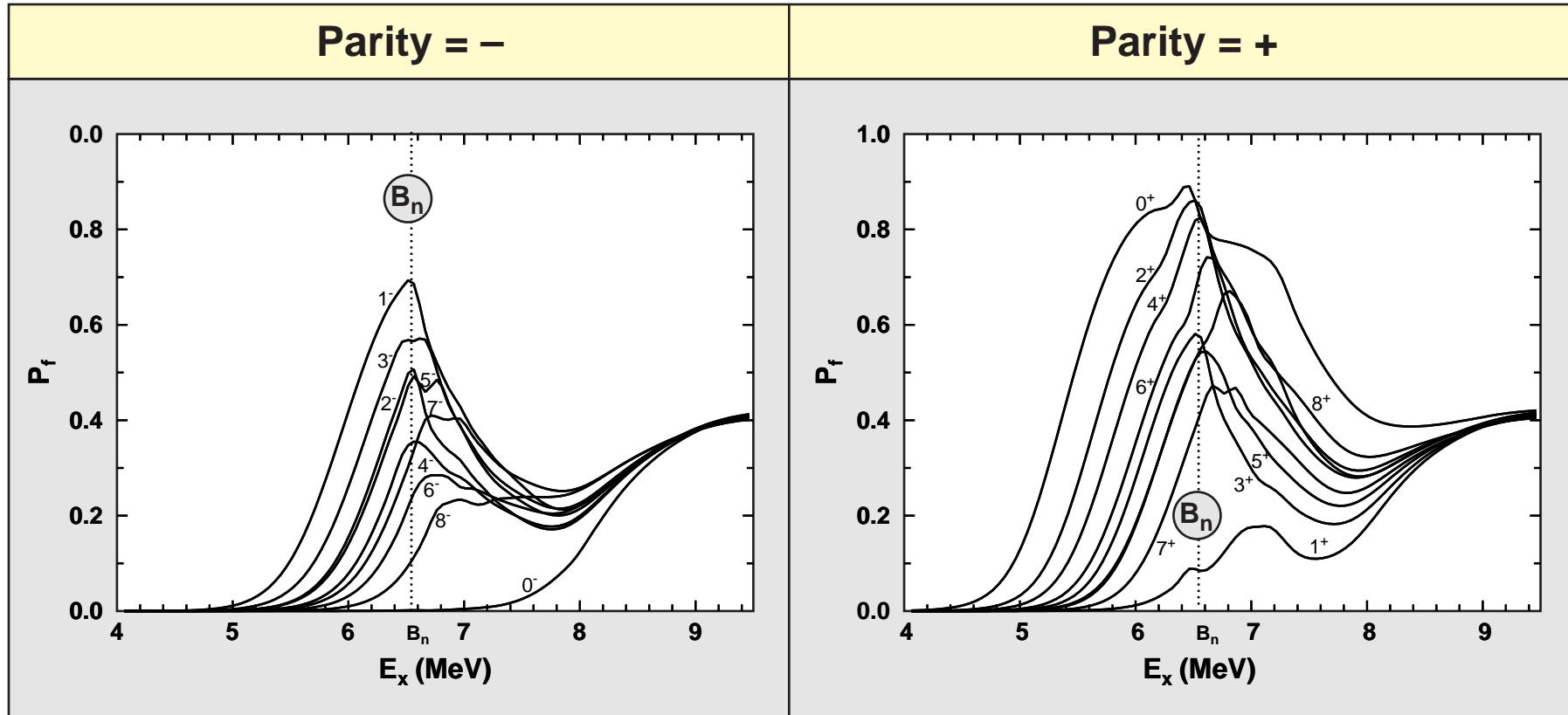


- Low-energy, low-spin fission requires low-spin transition states
- ^{235m}U has $J^\pi = 1/2^+$, ^{235}U has $J^\pi = 7/2^-$
- ^{235m}U requires $0^-, 1^+$, states for low-energy fission
- $0^-, 1^+$ transition states are scarce below 2Δ



$$\sigma_m/\sigma_g < 1 \text{ for } E_n < 2 \text{ MeV}$$

Fission probabilities for $n + {}^{235}\text{U}$

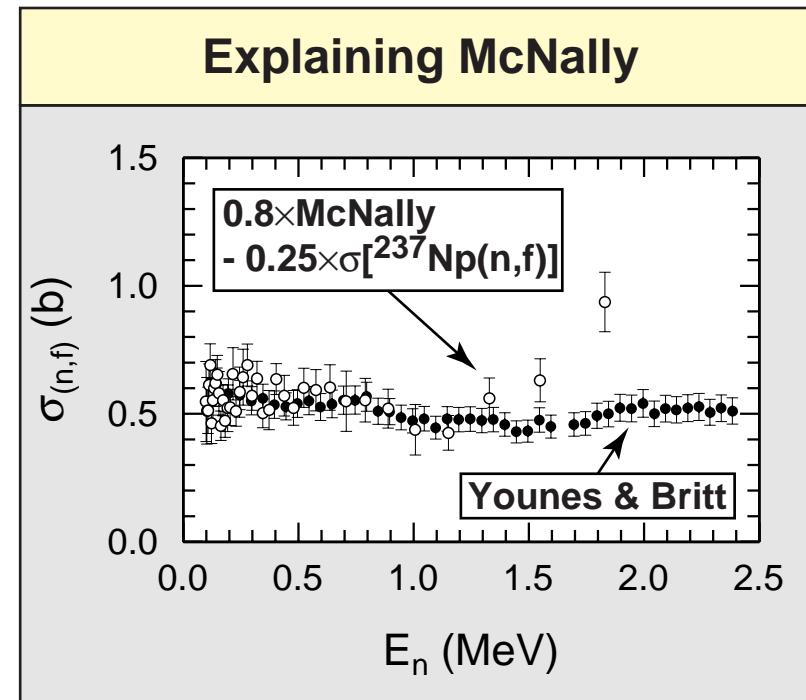
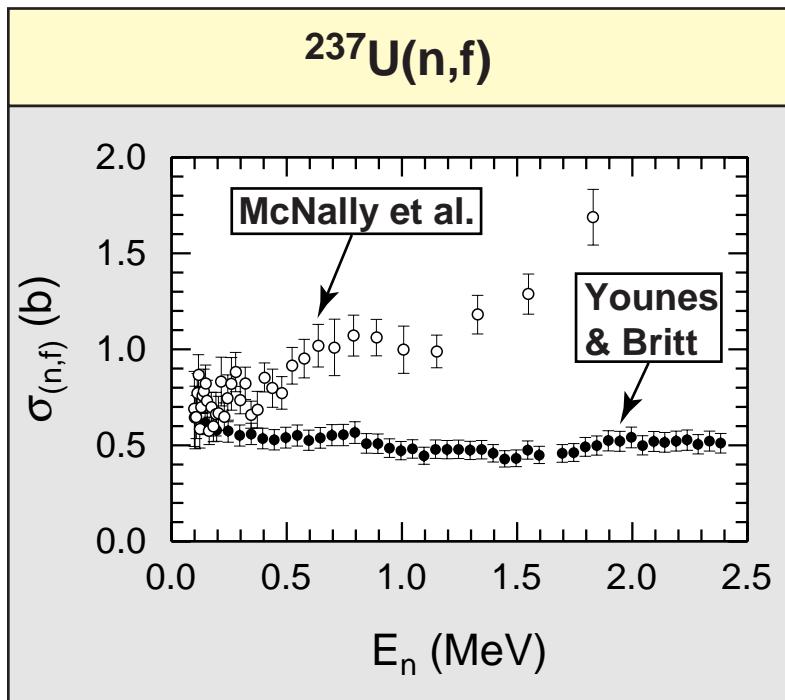


- 0^- , 1^+ probabilities are consistently lowest
- P_f is independent of J^π for large E_x : statistical effect

The case of $^{237}\text{U}(\text{n},\text{f})$

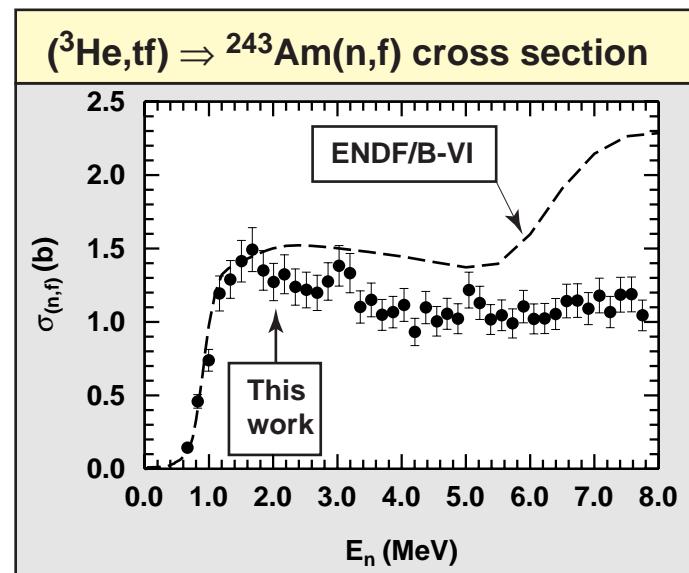
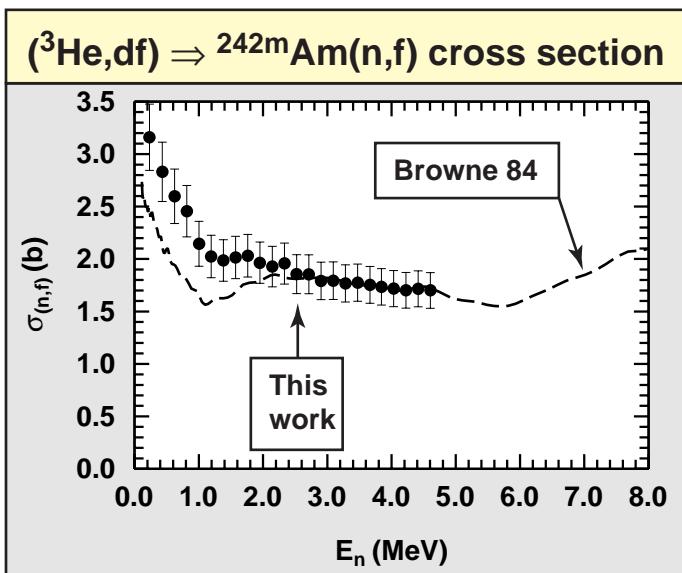
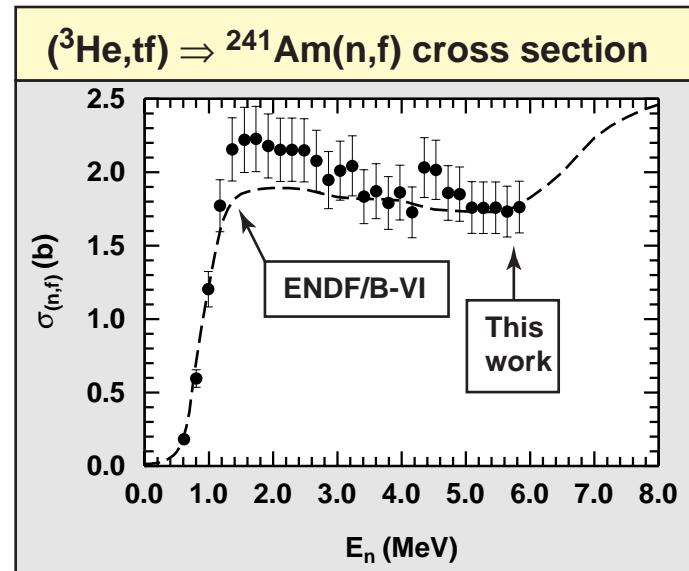
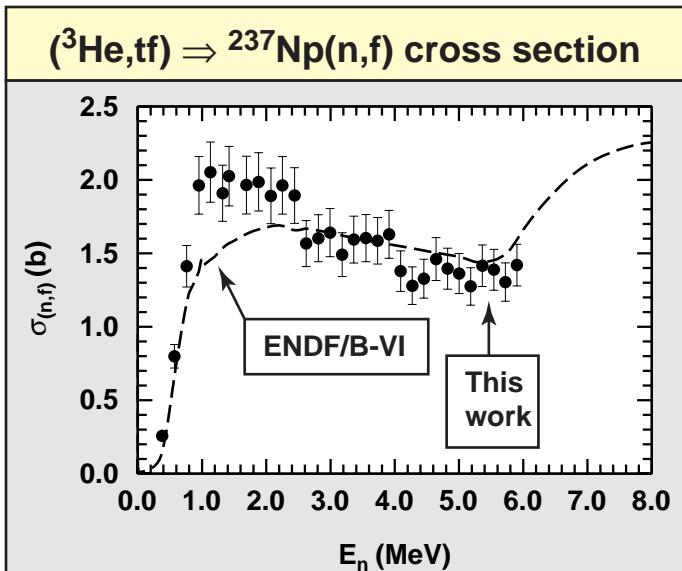


- Extremely difficult measurement: $T_{1/2}(^{237}\text{U}) = 6.8 \text{ days!}$
- Heroic effort by McNally *et al.* (1974): bomb shot
 - Data are suspect above $E_n \approx 400 \text{ keV}$ (^{237}Np contamination)
 - Data are inconsistent with critical-assembly measurements



- Our surrogate $^{237}\text{U}(\text{n},\text{f})$ cross section:
 - Is consistent with critical-assembly results (within 10%)
 - Explains McNally data as ^{237}Np contamination

Validation of the surrogate method for $(^3\text{He},\text{xf})$

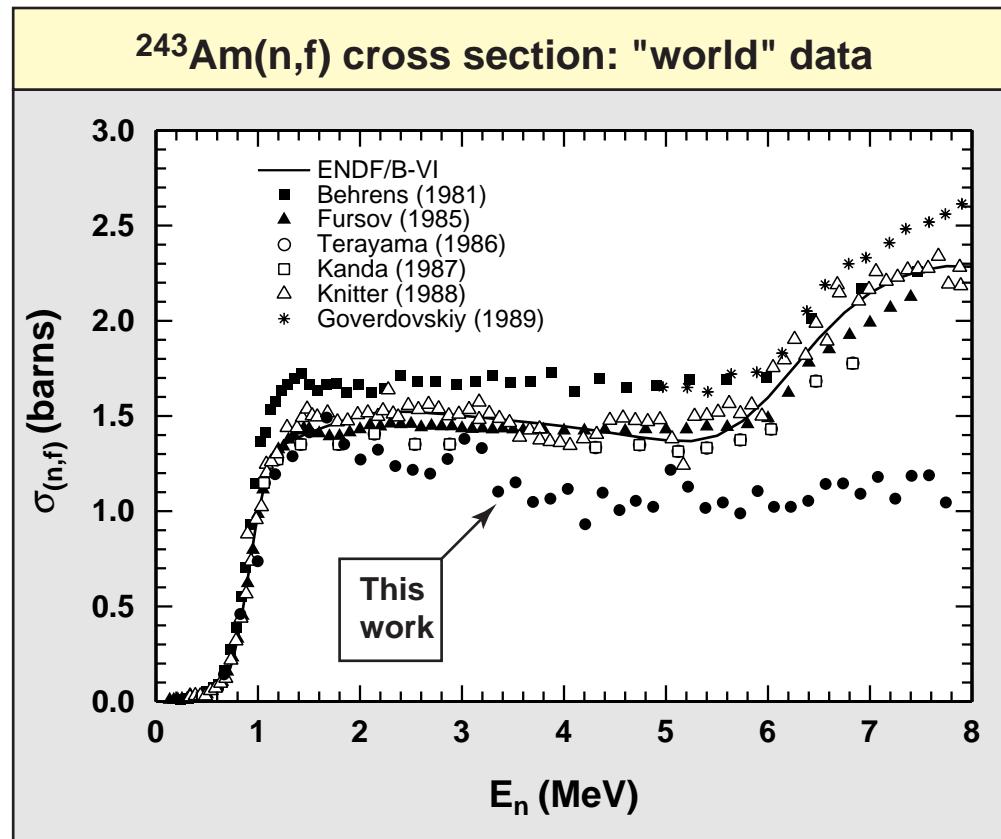


Overall, estimates agree to 10% above 1 MeV, except $^{243}\text{Am}(\text{n,f})$



The $^{243}\text{Am}(n,f)$ discrepancy

- Present estimate disagrees with (n,f) data



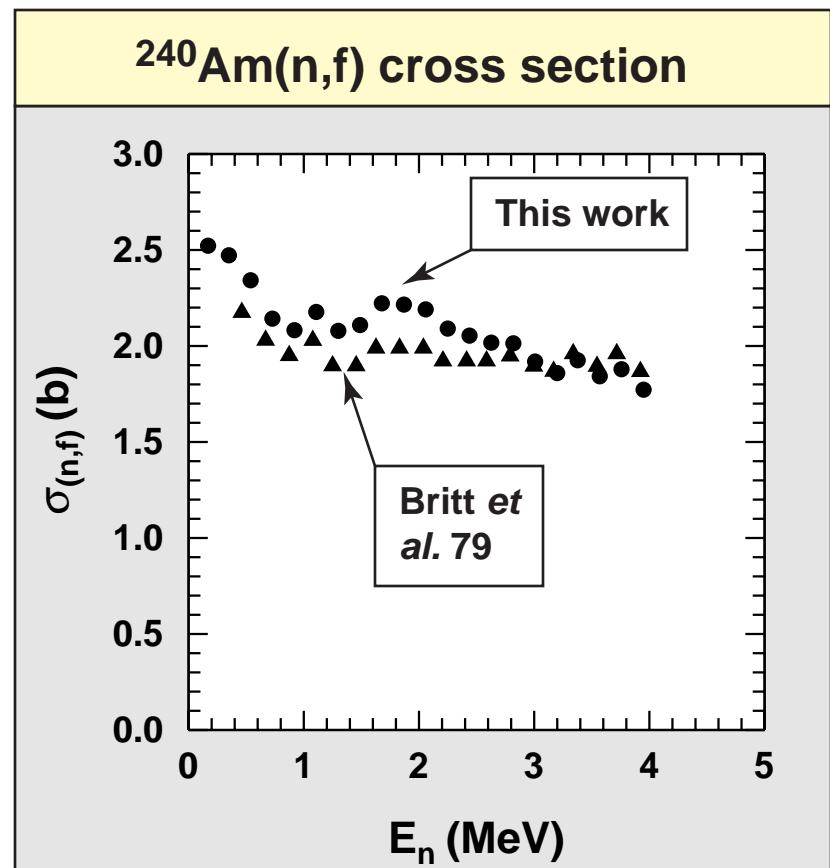
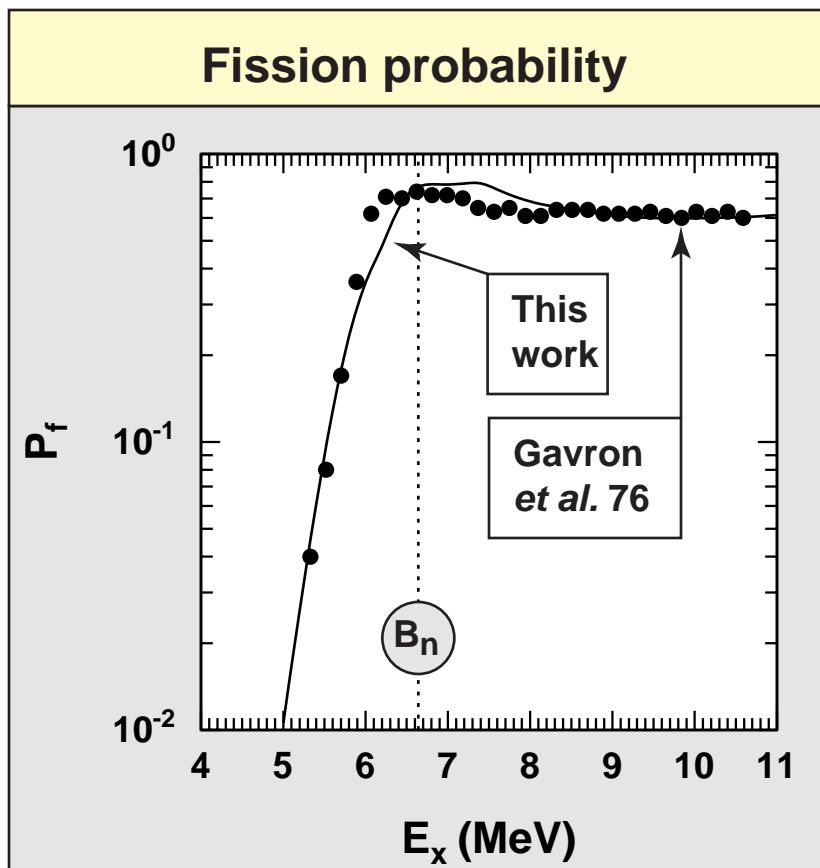
- Problem with ^{244}Pu target
 - Known tungsten contaminant
 - $^{244}\text{Pu}(^3\text{He},\text{tf}) \Rightarrow ^{243}\text{Am}(n,f)$
 - $^{244}\text{Pu}(^3\text{He},\text{df}) \Rightarrow ^{244}\text{Am}(n,f)$

Expect ~ 10% reliability for $E_n = 1\text{-}6$ MeV for most $(^3\text{He},xf)$ results
– but $^{243}\text{Am}(n,f)$ and $^{244}\text{Am}(n,f)$ estimates are less reliable

Application: $^{240}\text{Pu}(^3\text{He},\text{df}) \Rightarrow ^{240}\text{Am}(\text{n,f})$

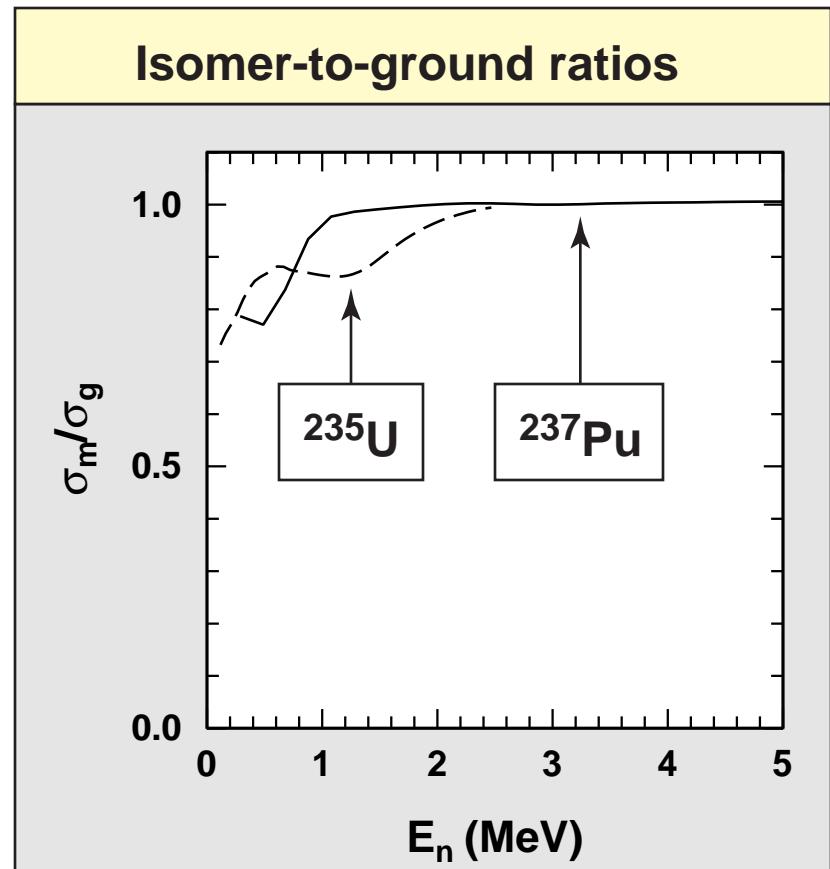
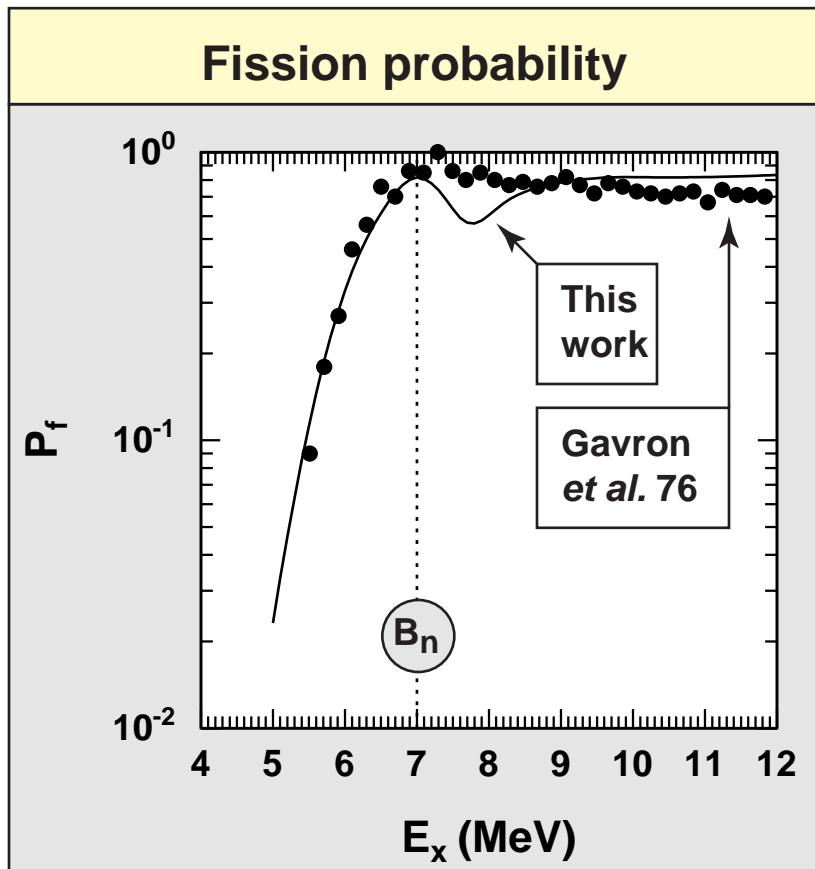


- $T_{1/2} \approx 51 \text{ h} \Rightarrow \text{Can't measure (n,f) any other way}$



Previous and current estimates are consistent

Application: $^{237}\text{Np}(^3\text{He},\text{df}) \Rightarrow ^{237,237m}\text{Pu}(\text{n,f})$

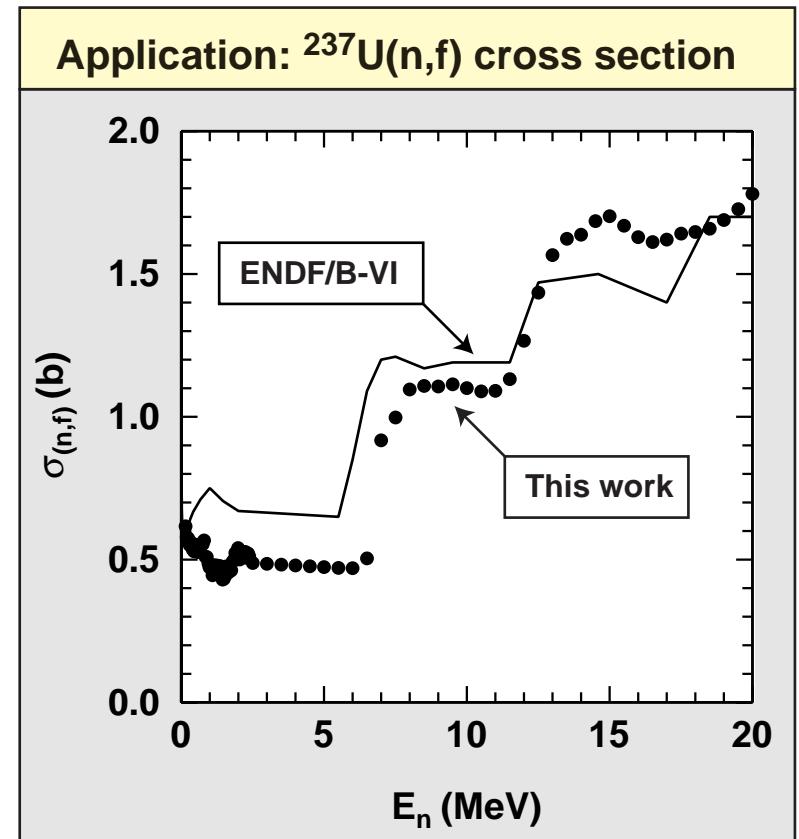
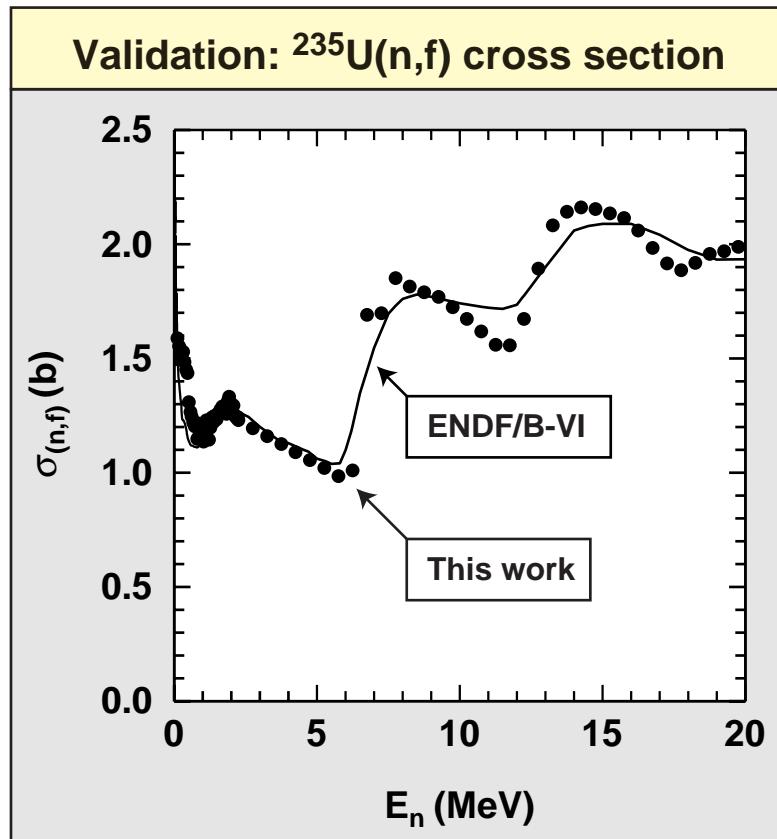


- ^{235}U and ^{237}Pu : similarities
 - both have $7/2^-$ ground and $1/2^+$ isomer
- Fission barriers somewhat lower for ^{238}Pu
- Neutron binding energy higher for ^{238}Pu



Actinide (n,f) up to $E_n = 20$ MeV

- Example: $^{237}\text{U}(n,f)$
 - "Daisy-chain" surrogate results for $^{237}\text{U}(n,0nf)$, $^{236}\text{U}(n,0nf)$, ...
 - Correct for CN-cross section depletion and pre-equilibrium at each stage



First cut: Younes et al., UCRL-ID-154194 (2003)
– Looks promising

Extending the surrogate technique to $E_n = 20$ MeV



Challenges:

- Level densities up to $E_x \approx 26$ MeV
 - consistent treatment of discrete and quasi-continuum states
 - better treatment of collective enhancements
- Pre-equilibrium effects in (n,xnf)
- Better treatment of spin transfer to highly-excited states
 - Energy dependence?
 - Wave functions?

Alternate approach:

- Surrogate data up to (equivalent) $E_n = 20$ MeV

Summary



- Extracted (n,f) cross sections for targets of
 - $^{231,233}\text{Th}$
 - $^{234,235,235\text{m},236,237,239}\text{U}$
 - $^{236,236\text{m},237,238}\text{Np}$
 - $^{237,237\text{m},240,241,243}\text{Pu}$
 - $^{240,241,242,242\text{m},243,244,244\text{m}}\text{Am}$
- Agreement with (n,f) standards
 - generally ~20% for $E_n < 1$ MeV, ~10% above
- Planned extension to $E_n = 20$ MeV